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THEORETICAL STUDIES OF OXYGEN-IODINE LASER MECHANISMS AND NEW LASER CONCEPTS

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This report summarizes theoretical studies of molecular dynamics, of spin-forbidden molecular transitions, of gain-dependent dispersions in excimer lasers, and of path-integral approaches applied to descriptions of quantum optical phenomena. It also summarizes two other studies, one of injection locking of arrays of lasers and another of detuning effects in laser instabilities.

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CONTENTS

I. INTRODUCTION

One of the research efforts of the Institute for Modern Optics (IMO) has focused on the physics and chemistry of the oxygen-iodine laser and on new laser concepts. It consisted of theoretical studies at the IMO, the experimental work carried out by M. Burrows at the Air Force Weapons Laboratory (AFWL), and the work of Y. Namba during his guest professorship at the University of New Mexico.

This report summarizes only the theoretical studies at the IMO, and begins with the research in the theory of chemical-laser molecular dynamics. The next area of research summarized by the report is an investigation of ways to break spin selection rules in order to achieve possible direct laser action with otherwise spin-forbidden transitions. The report continues with a summary of research progress in the area of new laser concepts, which includes studies of gain-dependent dispersion in eximer lasers and studies of path-integral techniques for treating quantum optical phenomena. The latter part of the report consists of a synopsis of studies of injection locking of phased arrays of lasers, and of detuning effects in laser instabilities.

II. MOLECULAR COLLISION THEORY

An ongoing research effort at the IMO aims at the development of molecular collision theories particularly suitable for studies of rate processes which govern chemical-laser mechanisms. The goal of this effort is to investigate the dynamics of molecular energy transfer at the level of state-to-state collisional exchanges of energy between atomic and molecular species present in the gain medium of given chemically pumped lasers. This type of research provides information about chemical-laser molecular dynamics at the level of details which are difficult to investigate experimentally.

Prior to the contract period of this report there was considerable progress at the IMO in developing the statistical quasi-classical (SQC) approach (Refs. 1 and 2) for calculating molecular collision dynamics. The SQC method relies on the Wigner distribution function to specify the quantum states of colliding molecules before and after collision, and on classical dynamics to describe the nuclear motions during collision. The SQC method is but one example among many others in theoretical physics which demonstrate the fundamental nature of the Wigner distribution function in descriptions of phase-space dynamics. Interest in the broader applicability of the Wigner distribution function in treatments of physical, chemical and optical phenomena continues at the IMO, and a major review article on this subject is near completion.*

* Hillery, M., O'Connell, R., Scully, M. O., and Wigner, E. P., "Distribution Functions in Quantum Physics," in preparation.

The SQC method in its present form applies only to inelastic molecular collisions whose nuclear dynamics are governed by a single electronic potential energy surface of the collision complex. That is, the SQC method at present is not capable of treating collisional exchange of energy between electronic and vibrational/rotational degrees of freedom. Such electronic-vibrational-rotational (EVR) energy exchange, however, happens to characterize many of the rate processes important to chemical lasers. This is particularly true for the chemical oxygen-iodine laser, which is a primary candidate for application of the theories under development at the IMO. For example, important EVR energy transfer processes in the gain medium of the chemical oxygen-iodine laser include: $O_2(^1\Delta) + I(^2P_{3/2}) \rightarrow O_2(^3\Sigma) + I^*(^2P_{1/2})$, leading to population of the lasing species $I^*(^2P_{1/2})$; $I^*(^2P_{1/2}) + O_2(^1\Delta) \rightarrow I(^2P_{3/2}) + O_2(^1\Sigma)$, $I^*(^2P_{1/2}) + I_2(^1\Sigma) \rightarrow I(^2P_{3/2}) + I_2(^1\Sigma)$, and $I^*(^2P_{1/2}) + H_2O \rightarrow I(^2P_{3/2}) + H_2O$, leading to depletion of the lasing species $I^*(^2P_{1/2})$; self-quenching of $O_2(^1\Delta)$, $O_2(^1\Delta) + O_2(^1\Delta) \rightarrow O_2(^3\Sigma) + O_2(^1\Sigma)$; quenching of $O_2(^1\Sigma)$ by H_2O, $O_2(^1\Sigma) + H_2O \rightarrow O_2(^1\Delta) + H_2O$; and $O_2(^1\Delta) + I_2(^1\Sigma) \rightarrow O_2(^3\Sigma) + I_2(A^3\pi_u)$ and $O_2(^1\Delta) + I_2(A^3\pi_u) \rightarrow O_2(^3\Sigma) + 2I(^2P_{3/2})$, which likely govern the production of iodine atoms.

The need for descriptions of nuclear dynamics on multiple electronic potential energy surfaces, so critical to chemical-laser molecular dynamics, has prompted an emphasis of research at the IMO on directly confronting the problem of EVR energy transfer. While approximate theoretical approaches such as the SQC method show much promise, the way of generalizing

them to account for intricate interplay between nuclear and electronic degrees of freedom is not immediately clear. One reason for this is the lack of a rigorous formulation of molecular collision theory which at the very outset accounts consistently for electronic, vibrational and rotational degrees of freedom, and energy exchange between them. Derivations of approximate treatments of the molecular dynamics of EVR energy transfer and tests of their accuracy require such a rigorous theoretical framework.

The prototype of EVR energy transfer involves an atom A and a diatomic molecule BC,

$$A^{*} + BC \rightarrow A + BC^{\dagger(\ddagger)} \qquad (1)$$

Here A* refers to Atom A in an electronically excited state while $BC^{\dagger(\ddagger)}$ refers to molecule BC in an excited vibrational/rotational (electronic/vibrational/rotational) state. Using electronic energy surfaces for NaH_2 from electronic structure calculations (at the Hartree-Fock level) of Botschwina, et al (Ref. 3), McGuire and Bellum (Ref. 4) performed in 1979 the first quantum-mechanical close-coupling calculations of the dynamics for collisional transfer of electronic energy from $Na^*(3p, {}^2P)$ to vibrational degrees of freedom of the H_2 molecule. Prompted by this work they began an investigation of the rigorous quantum-mechanical theory of EVR energy transfer. Bellum has continued this investigation and recently made a significant breakthrough in the analysis of the Hamiltonian for atom-diatom collision processes (Eq. 1) in the

context of molecular space-fixed (SF) and body-fixed (BF) reference frames. This analysis* follows from an investigation of forms of the molecular system Hamiltonian valid for rigorous quantum-mechanical treatments of inelastic atom-diatom collisions characterized by EVR energy exchange. The Hamiltonian assumes different forms depending on choice of independent coordinates which unambiguously specify the electronic and nuclear positions in the context of SF and BF reference frames. The Hamiltonian analysis focuses on the following four sets of independent coordinates: (1) a so-called "SF set", in which both electronic and nuclear positions are relative to the SF frame; (2) a so-called "mixed set", in which nuclear positions are relative to the BF frame while electronic positions are relative to the SF frame; (3) a so-called "BF set", in which both electronic and nuclear positions are relative to the BF frame; and (4) another "mixed set", in which nuclear positions are relative to the SF frame while electronic positions are relative to the BF frame. Practical considerations in accounting for electronic structure and non-adiabatic coupling of electronic states of the collision complex show that the forms of the Hamiltonian in the context of coordinate sets (3) and (4) above are most appropriate, respectively, for BF and SF treatments of nuclear dynamics in EVR energy transfer. These results constitute the first complete analysis of the Hamiltonian for a triatomic molecular

* Bellum, J. C., and McGuire, P., "Quantum-Mechanical Theory for Electronic-Vibrational-Rotational Energy Transfer in Atom-Diatom Collisions: Analysis of the Hamiltonian," submitted to J. Chem. Phys.

system and represent a significant step forward in the development of a rigorous scattering theory for EVR energy transfer which lends itself to actual calculations of the related collision dynamics. Analysis of related molecular system scattering wavefunctions of a prototype EVR energy transfer process (Eq. 1) will follow.

III. SPIN-FORBIDDEN TRANSITIONS AND RELAXATION OF SPIN SELECTION RULES

During the contract period of this report the research at the IMO included an analysis of the theory of spin-forbidden transitions. The goal was to assess possibilities of enhancing rates of transitions which are otherwise spin-forbidden. Such prospects, if feasible, would have far-reaching implications for chemical laser technology because they would allow direct lasing from metastable, energy-carrying molecules such as $O_2(^1\Delta)$. Such direct lasing would obviate the complications surrounding the numerous competing molecular rate processes which are characteristic of chemical laser mechanisms based on collisional energy transfer from metastable molecules.

One way of influencing rates of spin-forbidden electronic transitions is via application of intense, strongly inhomogeneous, external, magnetic fields. According to calculations, however, significant enhancement of such transition rates results only for magnetic-field inhomogeneities of the order of magnitude of 10^{12} gauss/cm. The requirement of magnetic fields of such enormous inhomogeneity places this scheme for increasing rates of spin-forbidden transitions well into the realm of impracticality. Application of external inhomogeneous magnetic fields represents only one way of relaxing spin selection rules characterizing metastable molecular states. Investigation of other more feasible schemes continues at the IMO.

IV. GAIN-DEPENDENT DISPERSION IN EXCIMER LASERS (Ref. 5)

The class of rare-gas halide lasers (e.g., XeF, KrF, and XeCl) has potential applications in high-power laser systems. These lasers involve transitions between rotational-vibrational levels of different electronic bands. Because of a broad and highly overlapping rotational structure within the gain profile, the effects of optical dispersion in rare-gas halide lasers may be sufficiently large to affect output-beam quality. For example, estimates, based on the assumption of overlapping Lorentzian lines with simplified line-spacing and line-strength assumptions, have produced phase shifts at the peak-gain wavelength in the range $-0.3\ g < \frac{d\phi}{dx} < -1.2\ g$, where g is the local loaded-power-gain coefficient. Since the gain across an amplifier cross section is not constant, one can envision sending into the amplifier a Gaussian beam and getting an output with an aberrated wavefront.

Research at the IMO in this connection consisted of a more sophisticated analysis based on semiclassical laser theory. This analysis gives the frequency dependence of the gain and anomalous dispersion analytically, and in applications to a particular laser transition of a XeF laser predicts a frequency within the gain curve where $d\phi/dx = 0$. The theory, however, is sufficiently general to be applicable to studies of the other rare-gas halide lasers.

According to the theory, the phase shift $d\phi/dx$ is maximum at the short-wavelength edge of the gain curve. At maximum gain, the numerical results give $d\phi/dx = -0.51\ g$. An important result, from the point of view of output-beam quality in a XeF

amplifier, is that $d\phi/dx = 0$ for operation at a wavelength that is detuned 1.2 Å to the red of the gain maximum. At this wavelength, the gain is reduced only by about 20%.

Noteworthy in the case of operation in an optical resonator (XeF oscillator) is the expectation that affects of mode pulling and mode pushing are not large. This is because the resonator has a greater effect in determining the phase of the intercavity laser field, and its stabilizing influence on the laser field is greater than the detrimental effects of the gain-induced dispersion. In the case of amplifier operation, however, this is no longer true, and problems with output-beam quality may result for operation at wavelengths where $d^2\phi/(dgdx)$ is small.

V. PATH-INTEGRAL APPROACH TO PROBLEMS IN QUANTUM OPTICS (Ref. 6)

Path integrals and the approximations to which they have led have found wide use in quantum field theory. The path-integral representation of the propagator allows one to see more clearly than does the standard operator approach the connection between the classical and quantum dynamics of a system. Derivations of semiclassical approximations then follow in a natural way. This aspect of path-integral approaches makes them particularly useful in molecular scattering theory (Ref. 7). So far, however, use of these techniques in quantum optics has been minimal.

Research into path-integral techniques at the IMO has led to development of some of the formalism which will facilitate their applications to certain phenomena in nonlinear optics. The types of problems to which these techniques apply are those in which the medium that interacts with the light can be described by a nonlinear susceptibility tensor. These include such processes as parametric amplification and harmonic generation. The interaction between the different modes is then described by products of various powers (depending upon the specific process) of the creation and destruction operators of the modes involved.

The type of path integral which we consider is not the one based on coordinate representations of the field which are typical of path-integral applications in quantum field theory. Path integrals based on representations of modes of the field in terms of coherent states lend themselves to treatments of problems in which only a few of these modes are important.

Hamiltonians expressed in terms of creation and destruction operators, and not the corresponding position and momentum operators, have as natural states the coherent states, which are eigenstates of the destruction operator. The calculation of the matrix element of the time development transformation between two coherent states then follows in terms of the coherent state path integral. This matrix element represents a type of propagator.

Out of this research has come discussion of various properties of the propagator, demonstration of its role in the calculation of quantities of interest in quantum optics, and derivation of formulas to calculate the propagator for single-mode systems with Hamiltonians at most quadratic in the creation and destruction operators. Aplication of these formulas has focused on calculation of the propagator for the case of second subharmonic generation when the pump field is classical. Generalization of these results permits calculation of the propagator for an N-mode system whose Hamiltonian is quadratic. This result provides the basis for investigation of the propagator for a parametric amplifier with a classical pump field.

VI. PHASE LOCKING OF LASERS BY AN INJECTED SIGNAL (Refs. 8-10)

The idea of using a phased array of lasers to obtain higher output power has recently received considerable interest. Basically, the scheme involves arranging a number of lasers in parallel and fixing the relative phases between the lasers by injecting into each laser part of the output of a stable laser. An advantage of this setup is that each laser in the array can be of a size that falls within construction constraints of current technology. Therefore one should not encounter any new problems involving fabrication and damage resistance of materials. The phase locking of the lasers causes the whole device to act as one spatially coherent source. Consequently, one expects a smaller focal volume and a higher power density in the far field than is the case with n randomly phased lasers. Whether this can be achieved in practice depends on precise locking of the phase of each laser in the array to a common master laser.

The equations describing a laser in the presence of an injected signal are

$$dE_L/dt = gE_L + \Delta_c T^{\frac{1}{2}} E_i \cos(\phi_L - \phi_i) \tag{2}$$

and

$$d\phi_L/dt = \nu_L - \Delta_c T^{\frac{1}{2}} (E_i/E_L) \sin(\phi_L - \phi_i) \tag{3}$$

where E_L and ϕ_L are the intracavity electric-field amplitude and phase, respectively, g is the loaded net gain, ν_L is the free-running laser frequency, Δ_c is the frequency spacing between the modes of the passive resonator, T is the transmission of the mirror on which the signal impinges, and E_i and ϕ_i are the amplitude and the phase, respectively, of the injected signal measured outside the resonator. According to Equation 2 frequency locking (i.e., $d\phi_L/dt = d\phi_i/dt \equiv \nu_i$) occurs when

$$E_i/E_L \geq |\nu_L - \nu_i|/(\Delta_c T^{1/2}) \tag{4}$$

The relative phase of the laser field in this case is

$$\phi_L - \phi_i = \arcsin\{[(\nu_L - \nu_i)/(\Delta_c T^{1/2})](E_L/E_i)\} \tag{5}$$

A phased array of lasers requires fixed values of $\phi_L - \phi_i$ for each laser in the array. According to the above equation, phase locking requires $\nu_L - \nu_i$ to remain constant. Fluctuations in the optical-resonator length due to mechanical vibrations or temperature variations, however, cause fluctuations in ν_L. Since the length changes in the lasers of the array are uncorrelated, the relative phases between lasers may be random even though they have the same frequency. An analysis to determine if an injected signal can lock the phase of a laser in the presence of these fluctations shows that, in the presence of an injected signal, the power spectral density is no longer Lorentzian. The narrowing of the spectral density corresponds to a stabilization of the phase of the laser field. In all

cases it is possible to dampen the phase noise so that the spectral density approaches that of the injected signal. For a free-running laser with a bandwidth of 1.5 MHz, calculation shows that the bandwidth is reduced to 15 kHz, i.e., by 2 orders of magnitude, when

$$i/I = 4 \times 10^{-4} \tag{6}$$

where $T = 0.01$ and $L_0 = 3m$. In practice, one can reduce the injected-signal strength necessary to maintain phase locking by stabilizing the laser-resonator length as much as possible.

VII. EFFECTS OF DETUNING ON SINGLE-MODE LASER INSTABILITIES (Ref. 11)

The stability of single-mode operation of high-power CW lasers was the topic of this work, which has possible aplication to laser systems such as the chemical oxygen-iodine and the HF lasers. Linear stability analysis shows the effects of detuning on single-mode instabilities in unidirectional ring lasers. According to the theory there are two regions of single-wavelength instability in homogeneously broadened lasers. The first is at line center, for which population pulsations are solely responsible, and the second is off line center where the unsaturated medium provides the required gain and anomalous dispersion. This research consists of numerical studies of four cases: homogeneous and inhomogeneous broadening, each with a single wavelength and with multiple wavelengths.

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